- **6**. The method according to claim **1**, wherein the step of preparing the gel from the fibroin solution comprises a gelling reagent containing phosphate ions.
- 7. The method according to claim 2, wherein the material is treated with calcium ions to form a fibroin-apatite before treating the material with the cross-linking agent.
  - 8. (canceled)
- **9**. The method according to claim **7**, wherein the calcium ions are provided by a solution of calcium chloride.
- 10. The method according to claim 9, wherein the material is washed with ethanol to remove excess calcium chloride and to convert the fibroin into a silk II state.
- 11. The method according to claim 10, wherein the material is dried after the washing step.
- 12. The method according to claim 2, wherein the cross-linking agent includes one or more of hexyl isocyanate (HMI), methyl isocyanate (MIC), hexamethylene di-isocyanate (HDI), methylene diphenyl di-isocyanate (MDI), toluene di-isocyanate (TDI) and isophorone di-isocyanate (IPDI).
- 13. The method according to claim 2, wherein the treatment with the cross-linking agent is carried out with substantially no fibroin swelling agents.
  - 14. (canceled)
- 15. The method according to claim 2, wherein the method comprises the further step of removing excess cross-linking agent from the material in one or more rinsing steps.
  - 16. (canceled)

- 17. The method according to claim 15, wherein the method comprises the further step of drying the material.
  - 18. (canceled)
- 19. The method according to claim 1, wherein the fibroin solution is a regenerated fibroin solution.
- **20**. The method according to claim **19**, wherein the regenerated fibroin solution is prepared by a method comprising treating silk or silk cocoons with an ionic reagent comprising an aqueous solution of monovalent cations and monovalent anions, the cations and anions having ionic radii of at least 1.05 Angstroms and a Jones-Dole B coefficient of between –0.001 and –0.05 at 25° C.
- 21. A method for the preparation of an implantable material for the repair, augmentation or replacement of bone from a regenerated fibroin solution, wherein the regenerated fibroin solution is prepared by a method comprising step of treating silk or silk cocoons with an ionic reagent comprising an aqueous solution of monovalent cations and monovalent anions, the cations and anions having ionic radii of at least 1.05 Angstroms and a Jones-Dole B coefficient of between -0.001 and -0.05 at 25° C.
  - 22-31. (canceled)
- **32**. An implantable repair, bone augmentation, or bone replacement material obtainable by the method according to claim **1**.
  - **33-47**. (canceled)

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